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B. FIELDS SEARCHED

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C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	Agarwal, G.S. Gupta, S.D., Interaction between surface plasmons and localized plasmons, Physical Review B, Vol.32, No.6, (1985), pages 3607 to 3611	1-54

☐ Further documents are listed in the continuation of Box C.☐ See patent family annex.

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Interaction between surface plasmons and localized plasmons

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A theoretical analysis of the experiments of Holland and Hall [Phys. Rev. B 27, 7765 (1983)] is given. The metal-island film is characterized by an effective anisotropic dielectric function. Formulas for reflection and transmission coefficients for an *anisotropic* layered structure are given. Calculated normal and non-normal reflectivities are in reasonable agreement with experiments. The results explain the observed shift in the surface-plasmon dispersion due to its interaction with localized plasmons.

I. INTRODUCTION

Recently Holland and Hall studied the interaction between the surface plasmons in a metal film and the localized plasmons in a Ag-island film.¹ The shift in the surface-plasmon dispersion was measured as a function of the separation between the two metal films.² The characteristics of the localized plasmons³⁻⁵ were studied by examining the normal-incidence reflectivity as a function of the wavelength. The purpose of this paper is to show that the observed features can be explained in terms of a rather simple model in which the island film is described by an effective dielectric⁵ function; though the results are very sensitive to the choice of parameters. Note that because of the structure of the islands, this film is anisotropic and thus in Sec. II we present the formulas for the calculation of the reflection and transmission coefficients from a layered structure where each layer can be anisotropic. In Sec. III we calculate numerically the reflection coefficient and obtain the changes in the dispersion characteristics of surface plasmons due to the interaction with localized plasmons. These results explain the broad features of the experiments of Holland and Hall. We also present ap-

proximate expressions for the normal-incidence reflectivities which show explicitly the connection with the imaginary part of the effective dielectric function of the island film.

II. REFLECTION FROM A MULTILAYER STRUCTURE WITH ANISOTROPY

The experiments of Holland and Hall use a multilayer geometry consisting of a prism, a Ag film, a spacer layer of LiF, a Ag-island film and air. The island film can be characterized by an effective dielectric function, which in general, is anisotropic. Hence we need to calculate the reflection and transmission coefficients of a multilayer structure⁶ with anisotropy. Such reflection coefficients can be calculated from the knowledge of the general structure of the electromagnetic field in an anisotropic medium.

In a uniaxial medium with dielectric tensor $\epsilon_t = \epsilon_{xx} = \epsilon_{yy}$, $\epsilon_n = \epsilon_{zz}$, $\epsilon_{\alpha\beta} = 0$, $\alpha \neq \beta$, and occupying the domain say $L_1 < z < L_2$, the fields are known⁷ to have the general form

$$\mathbf{E}(\mathbf{r}, \omega) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{i(ux+vy)} \left\{ \left[1, -\frac{u}{v}, 0 \right] (\epsilon_o^+ e^{isz} + \epsilon_o^- e^{-isz}) + \left[\left[-\frac{\epsilon_n}{\epsilon_t} \frac{ut}{(u^2+v^2)}, \frac{\epsilon_n}{\epsilon_t} \frac{vt}{(u^2+v^2)}, 1 \right] \epsilon_e^+ e^{izt} + (t \rightarrow -t, \epsilon_e^+ \rightarrow \epsilon_e^-) \right] \right\} du dv, \quad (2.1)$$

where

$$s^2 = \frac{\omega^2}{c^2} \epsilon_t - u^2 - v^2, \quad t^2 = \frac{\omega^2}{c^2} \epsilon_t - \frac{\epsilon_t}{\epsilon_n} (u^2 + v^2). \quad (2.2)$$

For each spatial component (u, v) , the general solution (2.1) involves four unknowns: $\epsilon_o^\pm, \epsilon_e^\pm$. The terms ϵ_o^\pm (ϵ_e^\pm) correspond to ordinary (extraordinary) waves. Here s and t represent the z component of the propagation vectors for these waves. The boundary conditions at each interface involve the continuity of the tangential components of \mathbf{E}

and \mathbf{H} . It turns out to be convenient to match the components

$$(uE_x + vE_y), (vE_x - uE_y),$$

$$(vH_x - uH_y), (uH_x + vH_y).$$

We can now study the reflection from a layered structure, using the usual matrix methods.⁶ The matching of the field components E_x, E_y, H_x and H_y will yield four equations for four unknowns which in turn will result in 4×4 matrices. It turns out that 4×4 matrices can be re-

duced to block-diagonal structure consisting of 2×2 matrices if we work with combinations

$$(uE_x + vE_y), (vE_x - uE_y), \\ (vH_x - uH_y), -(uH_x + vH_y).$$

From the structure (2.1) of the field, it is seen, for example, $uE_x + vE_y = 0$ for the ordinary wave; whereas $vE_x - uE_y = 0$ for the extraordinary wave. Thus the ordi-

nary and extraordinary waves get decoupled.

Assume that i th uniaxial medium occupies the domain from $z = L_{i-1}$ to L_i . Matching the boundary conditions at $z = L_i$ yields two equations

$$\begin{bmatrix} \epsilon_{\mu i+1}^+ \\ \epsilon_{\mu i+1}^- \end{bmatrix} = m_{\mu}^{i+1, i} \begin{bmatrix} \epsilon_{\mu i}^+ \\ \epsilon_{\mu i}^- \end{bmatrix}, \quad \mu = o \text{ or } e, \quad (2.3)$$

where $m_{\mu}^{i+1, i}$ are 2×2 matrices:

$$m_o^{i+1, i} = \begin{bmatrix} e^{is_{i+1}L_i} & e^{-is_{i+1}L_i} \\ s_{i+1}e^{is_{i+1}L_i} & -s_{i+1}e^{-is_{i+1}L_i} \end{bmatrix}^{-1} \begin{bmatrix} e^{is_iL_i} & e^{-is_iL_i} \\ s_i e^{is_iL_i} & -s_i e^{-is_iL_i} \end{bmatrix}, \quad (2.4)$$

and

$$m_e^{i+1, i} = \begin{bmatrix} -\frac{t_{i+1}\epsilon_{ni+1}}{\epsilon_{ii+1}} e^{it_{i+1}L_i} & \frac{t_{i+1}\epsilon_{ni+1}}{\epsilon_{ii+1}} e^{-it_{i+1}L_i} \\ \left[u^2 + v^2 + \frac{t_{i+1}^2\epsilon_{ni+1}}{\epsilon_{ii+1}} \right] e^{it_{i+1}L_i} & \left[u^2 + v^2 + \frac{t_{i+1}^2\epsilon_{ni+1}}{\epsilon_{ii+1}} \right] e^{-it_{i+1}L_i} \end{bmatrix}^{-1} \\ \times \begin{bmatrix} -\frac{t_i\epsilon_{ni}}{\epsilon_{ii}} e^{it_iL_i} & \frac{t_i\epsilon_{ni}}{\epsilon_{ii}} e^{-it_iL_i} \\ \left[u^2 + v^2 + \frac{t_i^2\epsilon_{ni}}{\epsilon_{ii}} \right] e^{it_iL_i} & \left[u^2 + v^2 + \frac{t_i^2\epsilon_{ni}}{\epsilon_{ii}} \right] e^{-it_iL_i} \end{bmatrix}. \quad (2.5)$$

The reflection coefficient can be evaluated in terms of these m matrices. For a N layer system, we can write (2.3) as

$$\begin{bmatrix} \epsilon_{\mu N+1}^+ \\ \epsilon_{\mu N+1}^- \end{bmatrix} = \left[\prod_{i=0}^N m_{\mu}^{i+1, i} \right] \begin{bmatrix} \epsilon_{\mu 0}^+ \\ \epsilon_{\mu 0}^- \end{bmatrix} \equiv M_{\mu} \begin{bmatrix} \epsilon_{\mu 0}^+ \\ \epsilon_{\mu 0}^- \end{bmatrix}. \quad (2.6)$$

Assuming incidence from the medium 0 (occupying domain $0 < z < -\infty$), the reflection coefficient becomes

$$\epsilon_{\mu 0}^- / \epsilon_{\mu 0}^+ = -\frac{(M_{\mu})_{21}}{(M_{\mu})_{22}} \equiv R_{\mu 0}. \quad (2.7)$$

On the other hand, for incidence from the medium $N+1$ (occupying domain $L_N < z < \infty$), the reflection coefficient is

$$\epsilon_{\mu N+1}^+ / \epsilon_{\mu N+1}^- = \frac{(M_{\mu})_{12}}{(M_{\mu})_{22}} \equiv R_{\mu N+1}. \quad (2.8)$$

Thus given the direction of incidence and the dielectric properties of each layer, the reflection coefficients can be calculated.

Note that the poles of (2.7) or (2.8) will determine the structure of the possible surface modes in the layered structure, where each layer can be anisotropic.

III. NUMERICAL RESULTS FOR REFLECTIVITY AND SHIFTS IN SURFACE PLASMON DISPERSION

We now use the formulas of Sec. II to calculate the reflectivity in the specific case used by Holland and Hall.

Assume that the Ag film occupies the domain $0 < z < d_1$, and that the spacer layer lies in $d_1 < z < d_2 + d_1$. These two layers are characterized by the isotropic dielectric functions ϵ_1 and ϵ_2 . The island film occupies the region $d_1 + d_2 < z < d_1 + d_2 + d_3$. The prism is on top of the silver film. The dielectric function of the Ag film has been taken from the work of Johnson and Christy.⁸ The dielectric function of the prism and LiF are chosen as $\epsilon_0 = 2.23$ and $\epsilon_2 = 1.9363$, respectively. The dimensions of the metal islands are much smaller than the wavelength and their shapes are generally ellipsoidal. Hence we use the effective-medium theory to write the approximate dielectric functions of the island film

$$\epsilon_{3\mu} = 1 + \frac{q(\epsilon_1 - 1)}{1 + (\epsilon_1 - 1)f_{\mu}}, \quad \mu = t, n, \quad (3.1)$$

where ϵ_1 is to be taken from Ref. 8.

The parameter q is the filling factor. The quantity f_{μ} depends on the geometry^{4,5} of islands. It is well known for various shapes. However, the value of f to be used in (3.1) gets modified due to a number of other effects³ such as due to the image dipoles, neighboring dipoles. Hence in what follows we leave f_{μ} as an open parameter and we try to estimate f_{μ} from the experimental curves of Holland and Hall. The effective thickness of the island film is given by

$$d_3 = (t_m)/q, \quad (3.2)$$

where t_m is the mass thickness.

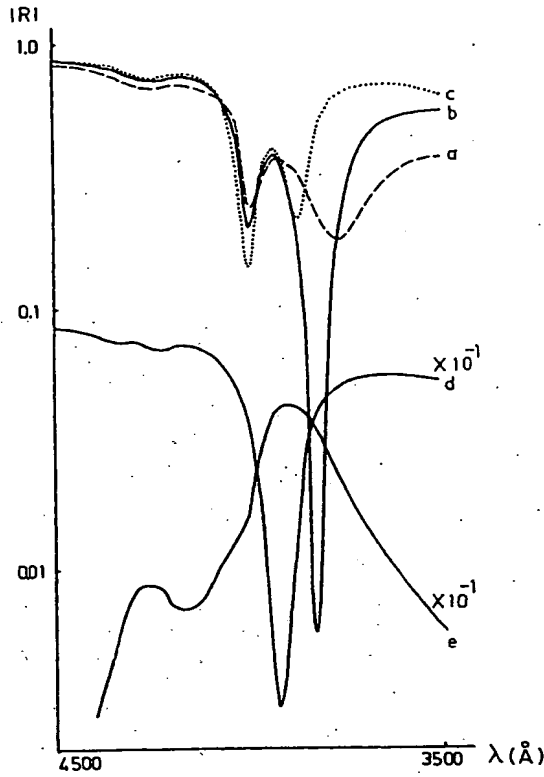


FIG. 1. The normal incidence reflectivity as a function of the wavelength of light incident from the island side of the structure. The parameters chosen are $\epsilon_0 = 2.23$, $\epsilon_2 = 1.936$, $f_1 = 0.19$, $q = 0.04$, $d_1 = 50$ nm, and the mass thickness of island film is 3 nm. Different curves are for different values of the spacer layer thickness $d_2 =$ (a) 40 nm (b) 55 nm, (c) 70 nm. For $d_2 = 55$ nm, curve d(e) gives the reflectivity for mass thickness 1 nm (continuous Ag film removed).

We first examine the behavior of the normal incident reflectivity $|R_n|$ as a function of wavelength of the light incident from the metal-island side of the layered structure. The reflectivity is calculated using Eq. (2.8) and by taking $u = v = 0$. From (2.2) it is clear that ϵ_1 enters the calculations and hence the reflectivity will depend on ϵ_1 . Thus the parameter f_1 will determine the normal incident reflectivity data. We have investigated $|R_n|$ for a wide range of parameters. The results are shown in Fig. 1. We find that the dip or the minimum on the blue side of 4000 Å is most pronounced for spacer-layer thickness ~ 55 nm.

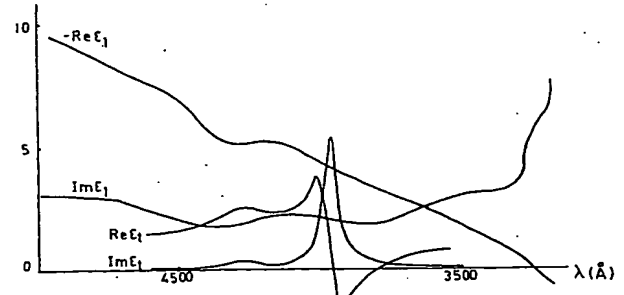


FIG. 2. Real and imaginary parts of the effective dielectric function of the island film as a function of wavelength. The real and imaginary parts of the dielectric function of the continuous Ag film (taken from Ref. 8) are also shown for comparison.

Reasonable values of the parameters f_1 and q are obtained by fitting the normal-incidence reflectivity curves so that for a given q , the position of the resonance occurs at the reported value. The parameter q is chosen such that the optimum behavior with respect to d_2 is obtained. For these values of the parameters the effective dielectric function of the island film is shown in Fig. 2. The two maxima in Fig. 2 correspond to the two absorption minima of Fig. 1. Figure 2 also shows for comparison the values of ϵ_1 obtained from the work of Johnson and Christy. Note that here we are considering a wavelength region in which Ag has a dielectric function whose real part is large and negative. In Fig. 1, we find that the absorption minimum corresponding to 4000 Å is split into two minima. This happens because small values of q make d_3 large if the mass thickness is kept constant. We have verified that for smaller mass thickness such a splitting is not there. This is also shown in Fig. 1 by the curve corresponding to mass thickness 1 nm. The extra feature is a consequence of the uniform-layer approximation for the island film. We now give an argument to explain the observed behavior in Fig. 1. We have to connect the expression for the reflectivity to the effective dielectric function of the island film. The reflection amplitude, as obtained from (2.8), has the explicit form

$$R = \frac{1-A}{1+A}, \quad A = \frac{m_{22} + (\epsilon_0)^{1/2} m_{21}}{m_{12} + (\epsilon_0)^{1/2} m_{11}}, \quad (3.3)$$

with

$$m_{22} = - \left[\frac{\epsilon_1}{\epsilon_r} \right]^{1/2} \sin(k_1 d_1) \cos(k_2 d_2) \sin(k_3 d_3) - \left[\frac{\epsilon_1}{\epsilon_2} \right]^{1/2} \sin(k_1 d_1) \sin(k_2 d_2) \cos(k_3 d_3) \\ - \left[\frac{\epsilon_2}{\epsilon_r} \right]^{1/2} \cos(k_1 d_1) \sin(k_2 d_2) \sin(k_3 d_3) + \cos(k_1 d_1) \cos(k_2 d_2) \cos(k_3 d_3), \quad (3.4)$$

$$m_{21} = -\frac{i}{(\epsilon_1)^{1/2}} \cos(k_1 d_1) \cos(k_2 d_2) \sin(k_3 d_3) - \frac{i}{(\epsilon_2)^{1/2}} \cos(k_1 d_1) \sin(k_2 d_2) \cos(k_3 d_3) \\ + i \left[\frac{\epsilon_2}{\epsilon_1 \epsilon_t} \right]^{1/2} \sin(k_1 d_1) \sin(k_2 d_2) \sin(k_3 d_3) - \frac{i}{(\epsilon_1)^{1/2}} \sin(k_1 d_1) \cos(k_2 d_2) \cos(k_3 d_3), \quad (3.5)$$

$$m_{12} = -i(\epsilon_1)^{1/2} \sin(k_1 d_1) \cos(k_2 d_2) \cos(k_3 d_3) + i \left[\frac{\epsilon_1 \epsilon_t}{\epsilon_2} \right]^{1/2} \sin(k_1 d_1) \sin(k_2 d_2) \sin(k_3 d_3) \\ - i(\epsilon_2)^{1/2} \cos(k_1 d_1) \sin(k_2 d_2) \cos(k_3 d_3) - i(\epsilon_t)^{1/2} \cos(k_1 d_1) \cos(k_2 d_2) \sin(k_3 d_3), \quad (3.6)$$

$$m_{11} = \cos(k_1 d_1) \cos(k_2 d_2) \cos(k_3 d_3) - \left[\frac{\epsilon_t}{\epsilon_2} \right]^{1/2} \cos(k_1 d_1) \sin(k_2 d_2) \sin(k_3 d_3) \\ - \left[\frac{\epsilon_2}{\epsilon_1} \right]^{1/2} \sin(k_1 d_1) \sin(k_2 d_2) \cos(k_3 d_3) - \left[\frac{\epsilon_t}{\epsilon_1} \right]^{1/2} \sin(k_1 d_1) \sin(k_2 d_2) \sin(k_3 d_3), \quad (3.7)$$

$$k_1 = \frac{\omega}{c}(\epsilon_1)^{1/2}, \quad k_2 = \frac{\omega}{c}(\epsilon_2)^{1/2}, \quad k_3 = \frac{\omega}{c}(\epsilon_t)^{1/2}. \quad (3.8)$$

The full expression is too complex. An understanding of the results of Fig. 1 can be obtained by analyzing the approximate form. We make a series of approximations—(a) Ignore the presence of the spacer layer, i.e., set $d_2 = 0$; (b) assume d_3 very small, so that we obtain reflectivity only to first order in d_3 ; (c) in the region of wavelength shown in Fig. 1 Ag has a large-negative real dielectric function, so we set $(\epsilon_1)^{1/2} \sim in_1$; (d) assume that the silver-layer thickness is large so that we can approximate $\sin(k_1 d_1), \cos(k_1 d_1)$ by $-(1/2i)e^{-ik_1 d_1}, \frac{1}{2}e^{-ik_1 d_1}$. Under these approximations we can write

$$R_0 \cong \frac{[(\epsilon_o)^{1/2} - 1] - ik_0 d_3 [\epsilon_t - (\epsilon_o)^{1/2}]}{[(\epsilon_o)^{1/2} + 1] - ik_0 d_3 [\epsilon_t + (\epsilon_o)^{1/2}]} \quad (3.9)$$

for the case when the uniform silver film is absent and

$$R_0 \cong \frac{[\epsilon_o^{1/2} - 1] - ik_0 d_3 [\epsilon_t - (\epsilon_o)^{1/2}]}{[(\epsilon_o)^{1/2} + 1] - ik_0 d_3 [\epsilon_t + (\epsilon_o)^{1/2}]} \quad (3.10)$$

if the silver film is present. On further simplifications, we find the following results for the reflectivity in the two cases:

$$|R_0|^2 \cong \left| \frac{(\epsilon_o)^{1/2} - 1}{(\epsilon_o)^{1/2} + 1} \right|^2 \left[1 + \frac{4k_0 d_3}{(\epsilon_o - 1)} \text{Im} \epsilon_t \right], \quad (3.11)$$

$$|R_1|^2 \approx 1 - \frac{4k_0 d_3}{(1 + n_1^2)} \text{Im} \epsilon_t. \quad (3.12)$$

The approximate results [(3.11) and (3.12)] show the remarkable connection between the reflectivity and the imaginary part of the dielectric function of the island film. Thus all the resonances (localized plasmons) of the ϵ_t of the island film will be reflected as peaks in $|R_0|$ and as dips in $|R_1|$. This explains the numerical results

of Fig. 1. It should be borne in mind that the numerical results are based on the exact result (3.3). The change of sign in the two cases can be understood as in the wavelength region of interest, Ag has negative real dielectric function. Thus the Ag film acts like a mirror, which accounts for phase change on reflection. We have thus shown that the localized plasmon resonances appear as peaks in the light reflected from the island film. However, such resonances appear as absorption resonances in presence of the additional Ag film.

Having obtained an estimate for q and f_t , we examine the coupling between surface plasmons and localized plasmons in the island film. Such a coupling is reflected in the reflectivity $|R_\phi|$ data as a function of the angle of incidence ϕ . The light is considered to be incident from the prism side. The calculation is done using (2.7), with $u = 0$, $v = \omega/c \sin \phi$, and with incident light polarization such that $\epsilon_o^+ \text{inc} = 0$, $\epsilon_o^- \text{inc} = 0$, i.e., incident light is p polarized. Figure 3 shows $|R_\phi|$ as a function of ϕ , both in

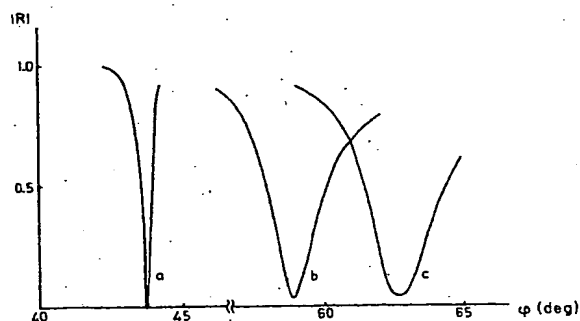


FIG. 3. The reflectivity R_ϕ as a function of angle of incidence for light incident from the prism side. The parameters are $f_n = 0.52$, $\lambda = 5145 \text{ \AA}$, and other parameters as in Fig. 1. (a) Ag, (b) Ag-LiF (55 nm), (c) Ag-LiF (55 nm) island.

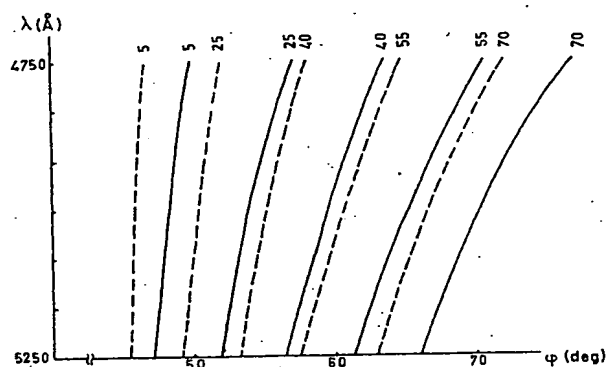


FIG. 4. The dispersion curves λ_ϕ vs ϕ . The solid (dashed) curves are in presence (absence) of the island film. Different curves are labeled by the thickness in nm of spacer layer. All other parameters are same as in Fig. 3.

the absence and in the presence of the island film. The parameter f_n is estimated from the sum rule. The reflectivity minimum λ_ϕ is investigated for a large number of wavelengths and the results are plotted in Fig. 4. We thus find changes in the surface-plasmon dispersion relation due to coupling with localized plasma resonances. Figure 4 gives λ_ϕ versus ϕ for a range of the thickness of the

spacer layer both in presence and absence of the island film. The shifts in the dispersion relation show an optimum behavior with regard to the thickness of the spacer layer, which is in conformity with the experimental results of Holland and Hall. It was also found that increasing q shifts the λ_ϕ versus ϕ curves to the right. Similarly increasing anisotropy (i.e., decreasing f_t) leads to the shifting of the dispersion curves to the left.

IV. CONCLUSIONS

In conclusion we have provided a theoretical model for the experimental observations of Holland and Hall. We have only tried to explain the broad features rather than detailed fitting with the data. The anisotropy of the island film is fully taken into account, though a simplified effective medium description of the island film is adopted. The model also assumes that the effects of substrate, neighboring islands can be accounted for by modifying the values of the parameters⁹ f_t and f_n . A better knowledge of f_t, f_n and an improved description of the island film are expected to yield results much closer to experiments.

ACKNOWLEDGMENT

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¹W. R. Holland and D. G. Hall, Phys. Rev. B 27, 7765 (1983); see also, W. R. Holland and D. G. Hall, Phys. Rev. Lett. 52, 1041 (1984). For a theoretical model for the results of this latter paper see D. Agassi and J. H. Eberly, Phys. Rev. Lett. 54, 34 (1985).

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⁹The island film may also have a distribution of the ellipsoid sizes and thus a better description can be obtained by averaging the effective dielectric function or the field distribution over the size distribution.